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Record of Issue/Revisions

ISSUE AUTHORIZATION DATE	EFFECTIVE DATE	REV. NO.	DESCRIPTION
Draft	10/24/2003	00-A	New document to establish TBD for the Y-12 National Security Complex – Occupational Internal Dose. Initiated by William E. Murray.
Draft	01/02/2004	00-B	Revised document in response to comments from OCAS and formal review on TBD for the Y-12 National Security Complex – Occupational Internal Dose. Initiated by William E. Murray.
Draft	03/05/2004	00-C	Revised document in response to additional comments from OCAS on TBD for the Y-12 National Security Complex – Occupational Internal Dose. Initiated by William E. Murray.
03/17/2004	03/17/2004	00	First approved issue. Initiated by William E. Murray.

ACRONYMS AND ABBREVIATIONS

ALI	Annual Limit on Intake
AMAD	Aerodynamic Median Activity Diameter
Bq	Becquerel
CEDE	Committed Effective Dose Equivalent
Ci	Curie
cpm	counts per minute
d	day
dpm	disintegrations per minute
DU	Depleted Uranium
EU	Enriched Uranium
g	gram
h	hour
HEU	Highly Enriched Uranium
HPGe	High Purity Germanium (detector)
ICRP	International Commission on Radiological Protection
in	inches
keV	kiloelectronvolt
L	liter
lbs	pounds
L _C	Critical Level: (commonly) 5% chance of a false positive
L _D	Detection Level: (commonly) 5% chance of a false negative
MDA	Minimum detectable activity
mg	milligram
min	minute
mL	milliliter
mm	millimeter
MPBB	Maximum Permissible Body Burden
MPC	Maximum Permissible Concentration
MPLB	Maximum Permissible Lung Burden
mrad	millirad
mrem	millirem
NBS	National Bureau of Standards (now National Institute of Standards and Technology)
nCi	nanocurie
NCRP	National Council on Radiation Protection and Measurements
NU	Natural Uranium
ORISE	Oak Ridge Institute for Science and Education
ORNL	Oak Ridge National Laboratory

PAL	Plant Action Limit
pCi	picocurie
PIPS	Passivated implanted planar silicon
ppb	parts per billion
ppm	parts per million
QF	Quality Factor
RU	Recycled Uranium
TPU	Total Propagated Uncertainty
TRU	Transuranic
y	years
µg	microgram
µm	micrometer

5.1 INTRODUCTION

From November of 1943, at the Y-12 Plant, now the Y-12 National Security Complex, uranium isotopes in various chemical and physical forms have been the primary contributors to internal radiation doses to workers. Other radionuclides have been handled at Y-12, and, at times, the uranium compounds processed may have contained impurities with radiological health implications. But the primary focus on internal dose control has been on uranium compounds and alloys over a wide range of ^{235}U enrichment. It is therefore appropriate to begin this section with an overview of concepts that apply broadly to uranium work over the history of Y-12. Key points developed in this and in subsequent subsections are summarized in Attachment A.

Two programs have been identified in the early years of Y-12 operations for which available information is currently insufficient to provide general guidance for internal dose reconstruction. The first program was associated with the research engineering and development group (RED), using a proton accelerating cyclotron in Building 9201-2, which became operational on November 11, 1950. Polonium isotopes and alpha airborne activity are the mentioned internal dose concerns. Later this same cyclotron was used to create neutron-deficient radionuclides. The second program is associated with plutonium and workers may have plutonium bioassay results in their records. When claim information indicates that a Y-12 worker was involved with research activities involving the calutron, cyclotron (accelerator), or fusion work or plutonium (except in the case of recycled uranium exposure which is addressed in this section), consideration must be given to possible exposure to radionuclides other than uranium.

5.1.1 Uranium Solubility in the Lung

Uranium compounds handled at Y-12 range from highly soluble to very insoluble. The primary hazard associated with soluble uranium compounds (depleted through low enrichment) has long been recognized to be nephrotoxicity. According to one reference, exposures to soluble compounds were monitored from the closing days of World War II by clinical tests of renal function and by fluorometric tests for uranium in urine (Sterner and Riley, 1946). However, no urinalysis data have been found prior to 1948 and this issue will have to be addressed in a revision of this document.

The lung was recognized as the primary organ of concern for the less soluble uranium compounds. During the greater part of the plant's history, control measures for less soluble compounds were guided by the lung and metabolic models presented in the International Commission on Radiological Protection (ICRP) Publication 2 (ICRP, 1960) or by similar predecessor models. These models related lung dose to uranium excretion in urine, particularly under chronic exposure conditions.

It is noteworthy that "Insoluble" material as described in ICRP Publication 2 would be classified as moderately soluble "Type M" material in the modern ICRP Publication 66 (1994) framework. It is also noteworthy that ICRP has recognized that some forms of uranium are more insoluble than initially believed. Figure 5-1 shows normalized lung retention for ICRP Publication 2 "Insoluble" material, ICRP Publication 30 (1979) "Class W" particles, Class "Q" material as described in the 1990's (Barber and Forrest, 1995), and Type M particles. The boxes on the diagram define the modern clearance Types F (fast), M (moderate), and S (slow) as a function of lung retention and time as illustrated in Annex D of ICRP Publication 71 (ICRP, 1995). The ICRP 2 insoluble model and the Class W models fit within the range of "Type M" material.

After *in vivo* lung counting was begun at Y-12 in 1961, it was recognized that a few workers had uranium lung burdens in considerable excess of the amount indicated by urinalysis. Writing in 1969, West and Scott noted that: “5 exposure cases noted in 1962 and 1963 showed chest burdens of enriched uranium which exhibited unexpectedly slow decreases after the persons were assigned to non-uranium activities.” (West and Scott, 1966 and 1969) The job description of all five individuals was “Process Operator” (Snapp, 2003).

Subsequent investigations suggested that three process combustion ashes from the enriched uranium (EU) foundry were the most likely exposure materials for these five cases (Steckel and West, 1966, p. 31). The particular thermal histories and particle size distributions of these process ashes were considered to be important factors leading to the low solubility in simulated lung fluid (Steckel and West, 1966, p. 30). Particle sizes tended to be relatively larger (0.86-1.6 μm) and the solubility to be lower for materials processed at higher temperatures.

While the exceptional cases with unusually protracted lung clearance are important, it is more important to note that, for the vast majority of individuals, lung clearance took place in approximate accordance with the ICRP Publication 2 (1960) “Insoluble” model, which fits within the current “Type M” framework.

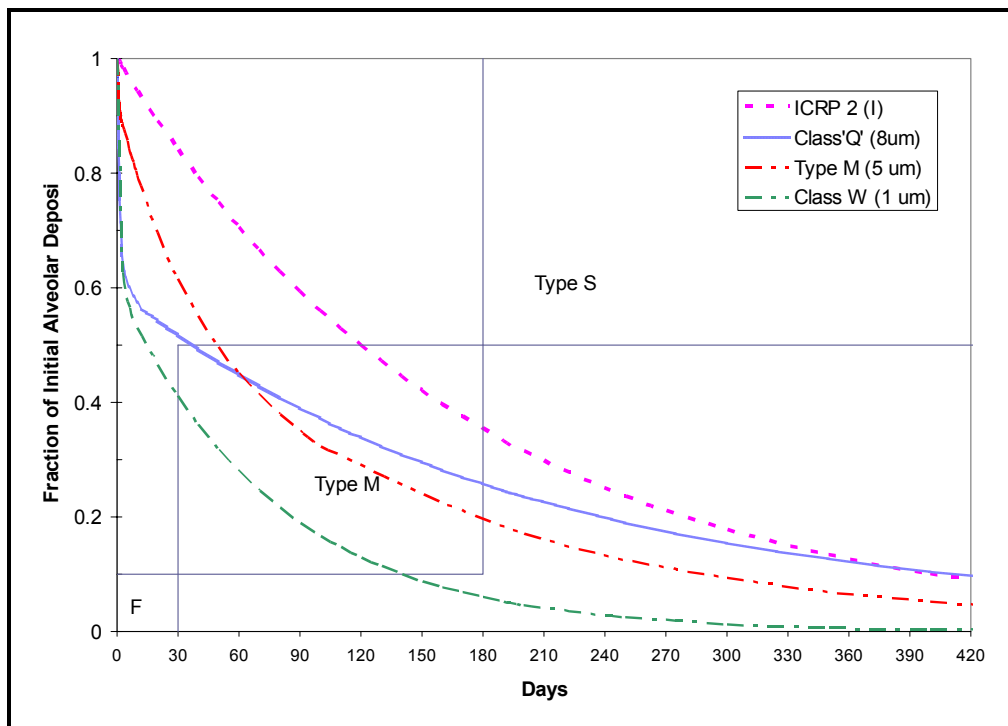


Figure 5-1. Historical solubility types meeting “Type M” definition.

In 1964, a composite urinary excretion curve was described for 157 workers who had been removed from uranium work because of high uranium urinalysis results (Scott, 1964). This empirical composite curve is shown in Figure 5-2, together with expected excretion curves for Types F, M, and S uranium. The latter curves are derived from 5 micrometer (μm) Aerodynamic Median Activity Diameter (AMAD) excretion curves (Potter, 2002). They were normalized to match the Scott curve at day 200 post removal from uranium work areas. The empirical excretion (Scott) curve matches the Type M curve much better than the Type F or Type S curves. Indeed, the Scott curve fits the Type M curve quite well over most of the 420-day time range. These results are consistent with the subsequent

conclusions of Rucker et al. that Class W is the appropriate default for uranium at Bechtel Jacobs facilities, including Y-12 (Rucker et al., 2001). Exceptions to this default included “high-fired” uranium oxides and compounds formed by the slow oxidation of uranium metal.

Beginning in September 1994, the plant was placed in a stand-down mode. This effectively stopped all routine work (and chronic exposure potential) in the process areas. Only minimal walk-throughs and area checks continued during the stand-down period which lasted until August 1998. The stand-down significantly influenced the available source term for exposures. Prior to stand-down, the Y-12 program default modeling assumption was Class Q (90% Super W, 10% Y). During the stand-down period, the Y-12 program default assumption of chronic exposure was modified to assume acute exposures occurring at the midpoint of a quarterly sampling frequency. Also, during this 4-year period, no wet chemical operations were performed. Thus, no soluble component was being produced and materials that were stopped in process were allowed to sit and oxidize which provided a larger component of Type S material as an exposure source. In August 1998, only partial resumption of activities occurred – wet chemical operations were not restarted and were still not restarted as of October 2003. The predominant material encountered after partial restart in August 1998 is uranium oxide, which fecal sampling has shown to be more consistent with 100% Type S material. Based on the changing workplace conditions after partial restart in 1998, the default assumptions were modified to return to chronic exposure but to use type S.

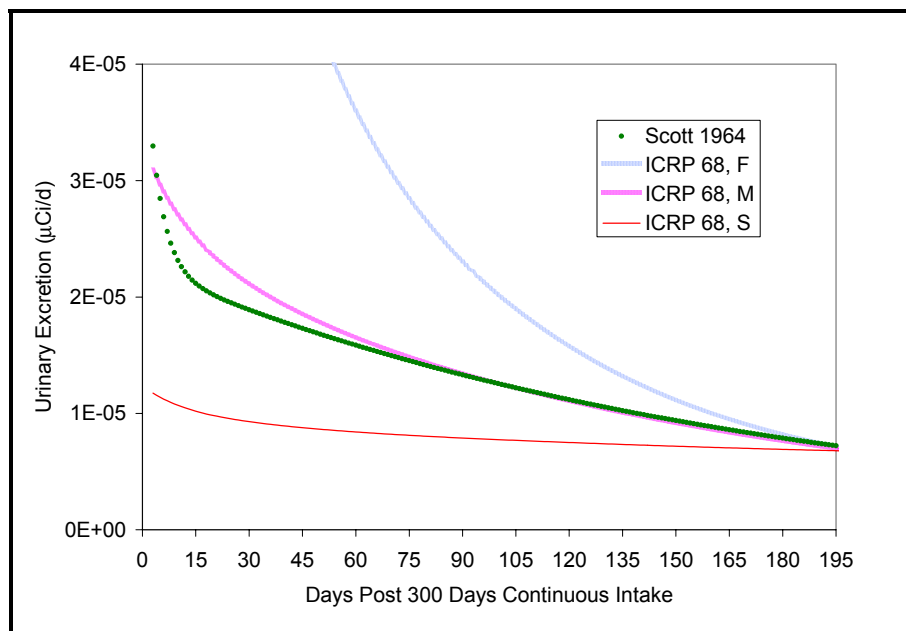


Figure 5-2. Empirical and theoretical uranium excretion after uranium restriction.

For a workplace as varied as Y-12, it is clear that no single solubility or particle size will apply to all workers. Furthermore, accurate assignment of the uranium lung clearance type to a given bioassay result was considered virtually impossible because of uncertainties regarding chemical form and limitations of the personnel-tracking system (Barber and Forest, 1995, p. 669). As of 2001, Y-12 implemented a Radiation Work Permit (RWP) tracking system that tracks the location of workers, their type of work, and the type of bioassay needed (see ORAUT-TKBS-0014-2, 2003, pp. 19 ff.).

Exposure to Type M material from 1948 to July 1998, appears to be the more likely absorption type. After July 1998, exposure to absorption Type S material is more likely. However, the absorption type may be based on the monitoring data and/or claimant favorable assumptions.

5.1.2 Particle Size Information

Several particle size studies in uranium process areas have been conducted at Y-12 (e.g., Y-940, 1953; Steckel and West, 1966; Barber and Forest, 1995; and BWXT Y-12, 2003b). For different times and different processes, reported particle sizes ranged from less than one to over 10 μm (physical). Steckel and West (1966) reported a positive correlation between uranium octoxide particle size and process temperature. Barber and Forest (1995) used an 8 μm AMAD based on particle size measurements as the basis for the "Class Q" dosimetry system used in the 1990s. (The "Class Q" system is further described as 10% Class Y and 90% modified Class W. The modification consists of increasing the Class W 50 day compartment to 120 days.) In terms of lung deposition and retention, the 8 μm AMAD Class Q material is closer to 5 μm AMAD Type M material than to 1 μm AMAD Class W material, as shown in Figure 5-3.

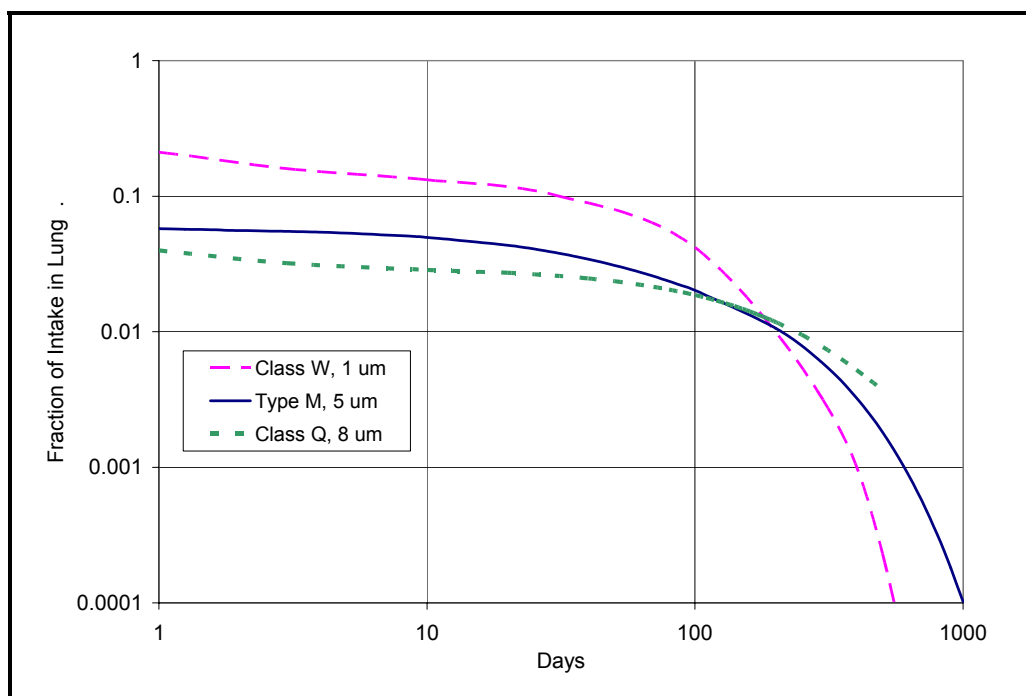


Figure 5-3. Lung deposition and retention of Class W, Class Q, and Type M uranium. (Particle sizes specified are Aerodynamic Median Activity Diameter (AMAD) in micrometer [μm].)

Since 2000, Y-12 has implemented the latest guidance provided by the ICRP including the ICRP Publication No. 66 (ICRP, 1994) lung model and ICRP Publication No. 78 (ICRP, 1998) methods and models (BWXT Y-12 2003a, p. 89). This is the justification for the change to the 5- μm AMAD as the default particle size. Y-12 uses the newer weighting factors proposed by the ICRP in its 1990 recommendations (ICRP, 1991). For Y-12 dose reconstructions, the default is a 5- μm AMAD particle size distribution.

5.1.3 Uranium Alpha Activity as a Function of Enrichment

At Y-12, uranium enrichments range from depleted uranium (DU) [<0.2 weight percent ^{235}U] to highly enriched uranium (HEU) [> 90 weight percent ^{235}U]. The potential dosimetric complexity is simplified by the similarity of the dose conversion factors for the most important uranium isotopes, and by the quantities measured in the *in vitro* bioassay and chest counting programs.

For work with enriched compounds and alloys, urinalysis programs after 1950 reported either total uranium alpha counts or isotopic uranium results, both of which reflect the dosimetric potential of the isotopes generally handled. Early *in vivo* chest counting results were reported either as micrograms (μg) of ^{235}U or milligrams (mg) of ^{238}U , based on whether an exposure was believed to be from enriched, natural, or depleted uranium. Based on these reporting techniques, the following assumptions may be made regarding data interpretation for the time period 1950 through 1989 for urinalysis and 1961 to 1989 for lung counting.

For lung counts a combination of the information in the Type Analysis and Material Type reporting fields can be used to determine whether the count was believed to be due to natural uranium (NU) or DU. For pre-1972 records, a Type Analysis of 1 and a Material Type of 2 or 3 indicate DU, while a Material Type of 7 indicates NU. For post-1972, a Type Analysis of 4 and a Material Type of 2 or 3 indicate DU, while a Material Type of 7 indicates NU.

At Y-12, it is current practice to assign dose by applying the dose conversion factor for ^{234}U to all inferred uranium intakes. This practice is slightly conservative for EU; the resulting overestimate is less than 1% of the effective dose. For DU, the use of the ^{234}U dose conversion factor leads to an overestimate of 16% for Type S uranium and 20% for Type M uranium (Eckerman and Kerr, 1999, p. 16). Information regarding the forms of uranium handled at Y-12 has been taken from Section 2, Site Description, Tables 2.4-1 and 2.4-2 (ORAUT-TKBS-0014-2, 2003), and is summarized below. If sufficient information regarding work locations/work activities is provided, the enrichment information in Table 5-2 may be used instead of the default information provided in Table 5-1.

Table 5-1. Enrichment assumptions to be used for dose assessment for various analytical techniques.

Analytical Technique (Urinalysis 1948-present) (Lung count 1961-1989)	Measurement Information	Default Enrichment Assumption
Urine by fluorometry (usually for an individual who worked in areas with natural/depleted uranium; prior to 1950, also used for workers in enriched uranium areas).	Alpha dpm can be calculated from mass.	Prior to 1950, 93% enrichment. After 1949, natural uranium.
Urine by gross alpha counting (usually for an individual who worked in areas with enriched uranium).	Alpha dpm is the sum of all uranium alphas.	93% enrichment.
Urine by alpha spectrometry (10/1989 to present).	Alpha dpm d^{-1} is reported isotopically and total uranium is determined by summing isotopic results.	
Lung Count	μg U-235	93% enrichment of 93%.
Lung Count	μg U-238	Natural uranium.

5.1.4 Temporal Pattern of Uranium Exposures

As a rule, routine uranium exposures at Y-12 were considered to be of a chronic nature. For example, in 1958, Patterson stated “. . . our interpretation of urinalysis results and our assignment of internal dose assume an exposure under equilibrium conditions of intake and elimination.”

Table 5-2. Summary of uranium enrichments by building and time period.

Building	1943-1948	1949-1951	1952-1963	1964-1995	1996-2003
9201-1	Uranium enrichment to 30%	uranium enrichment to 30%	Uranium salvage operations-all forms possible	Fusion energy research operations	n/a
9201-2	uranium enrichment to 30%	uranium enrichment to 30%	COLEX Lithium/Hg enrichment operations	Development/research operations	ORNL
9201-3	uranium enrichment to 30%	uranium enrichment to 30%	Uranium salvage operations-all forms possible	Fusion energy research operations	n/a
9201-4	uranium enrichment to 30%	uranium enrichment to 30%	COLEX Lithium/Hg enrichment operations	Engineering and administrative facilities	Awaiting D&D
9201-5	uranium enrichment to 30%	uranium enrichment to 30%	COLEX Lithium/Hg enrichment operations	Normal/depleted uranium	Normal/depleted uranium
9202	All forms handled	All forms handled	Uranium process development & improvement operations-primarily normal and depleted uranium	Uranium process development & improvement operations-primarily normal and depleted uranium	Technology Development – primarily normal and depleted uranium
9203	All forms handled	All forms handled	Recovery and salvage operations-primarily normal and depleted uranium	Y-12 production development and research operation-primarily normal and depleted uranium	Technology Development
9204-1	uranium enrichment to 95%	uranium enrichment to 95%	Stable isotope separation operations	Fusion energy research operations	ORNL facility, however BJC occupies office space
9204-2	uranium enrichment to 95%	uranium enrichment to 95%	Uranium assembly operations	Uranium assembly operations	Uranium assembly operations
9204-3	uranium enrichment to 95%	uranium enrichment to 95%	Stable isotope (e.g., copper) separation operations	Stable isotope (e.g., copper) separation operations	ORNL facility
9204-4	uranium enrichment to 95%	uranium enrichment to 95%	ELEX Lithium/Hg pilot-scale operations	Normal/depleted uranium	Depleted uranium
9206	All forms handled	All forms handled	Uranium chemical processing and metal production operations-primarily normal and depleted uranium	Uranium chemical processing and metal production operations-primarily normal and depleted uranium	Currently undergoing deactivation.
9207	Normal and slightly enriched	Normal and slightly enriched	Maintenance/salvage operations	ORNL biological research Operations	ORNL facility, mostly vacated.
9211	Normal and slightly enriched	Normal and slightly enriched	Recovery and salvage operations-primarily normal and depleted uranium	ORNL biological research operations	ORNL biological research operations
9212	All forms handled	All forms handled	UF ₆ conversion, chemical operations, and weapon production operations-primarily enriched uranium	Chemical operations and weapons production operations-primarily enriched uranium	Chemical operations and weapons production operations
9215	--	--	Enriched uranium	Enriched uranium	Enriched uranium
9998	--	Normal uranium	Depleted uranium	Depleted uranium	Depleted uranium

(Patterson, 1958, p. 58) During the stand down period from September 1994 until August 1998, acute exposures should be assumed to be the more likely mode of exposure. After this time, while complete equilibrium is not expected in modern internal dosimetry models, the presumption of chronic exposure conditions for uranium remains in place, e.g., "The most likely exposure potential for uranium work at the Y-12 Complex is chronic in nature." (BWXT Y-12, 2003a, p.107).

5.1.5 Other Radionuclides of Concern

In addition to ^{234}U , ^{235}U , and ^{238}U , the following radionuclides were identified in the Y-12 Technical Basis Document for Internal Dosimetry (BWXT Y-12, 2003a):

- ^3H , ^{90}Sr , ^{99}Tc
- ^{228}Th , ^{232}Th ,
- ^{232}U , ^{233}U , ^{236}U
- ^{238}Pu , ^{239}Pu , ^{241}Pu
- ^{237}Np , ^{241}Am

Radionuclides that could interfere with *in vivo* analysis of uranium and thorium were quantified so that their effects on the spectra could be taken into account:

- ^{40}K , ^{137}Cs

Other radionuclides addressed include:

- ^{60}Co and $^{95}\text{Zr}/^{95}\text{Nb}$ for organizations outside Y-12, (Cofield, 1961)
- ^{210}Po

Many of the radionuclides discussed above are accompanied by progeny in various stages of equilibrium. In addition, operators of the *in vivo* analysis equipment reviewed accumulating spectra for interferences from medical radioisotopes, such as ^{131}I .

Uranium has always been the dominant contributor to collective internal dose at Y-12. Monitoring for other radionuclides has been performed on a limited basis. The calculated uranium intakes can be used in conjunction with the ratios shown by building or area in Table 5-3 to determine the intake of the non-uranium radionuclide. The ratios are based on smear studies used to evaluate the relative activities of radionuclides in surface contamination. Survey locations were identified through research of historical documents and the results of interviews conducted with current and former employees who were familiar with non-U radionuclide use at Y-12 (BWXT Y-12, 2001, p. 3).

The U alpha:non-U ratios shown in Table 5-3 are from the current Internal Dosimetry monitoring program (BWXT-Y12, 2003a, pp. 99 ff), based on an assessment in 2001, and from results in the non-U hazard assessment performed at Y-12 in 1997 (LMES, 1997). The data from the latter study are identified with an asterisk (*). The radionuclides included in the non-U sum are the total activity of Th-228, Th-230, Th-232, Pu-238, Pu-239/240, Np-237, and Am-241. The tables identify the most restrictive non-U radionuclide based on ICRP 68 (1994a) inhalation dose conversion factors and the activity of the radionuclides. Thorium and plutonium were assumed to be Type S and americium and neptunium were assumed to be Type M for the comparison.

The actual laboratory results from the smears, as shown in Table 5-3, are likely to be representative of the materials, e.g., non-U radionuclides, that one would encounter during current activities, and not as a result of newly-introduced operations or processes involving non-U radionuclides. The values are believed to be a reasonable indicator, but are not necessarily an overestimate of the ratio of non-U radionuclides. The details on the development of the numbers in Table 5-3 as well as the actual smear results used to determine the U Alpha: non-U ratios are discussed in BWXT Y-12, 2001 and

LMES, 1997. The non-U intake can be inferred from the calculated U intake based on these ratios, assuming that the non-U intake is attributable to the most restrictive radionuclide detected in the smear results for the area of interest.

Table 5-3. Uranium to non-uranium ratios by building (LMES, 1997; BWXT Y-12, 2001).

Most Restrictive Non-U Radionuclide	Building / Area ¹	U Alpha: Non-U Alpha Ratio ²
Th-228	81-22, Land-Sea, Items, skid COMPOSITE	394
Th-228	9201-5, 1st floor, Th chip cleaning COMPOSITE	4
Th-228	9201-5, 1st floor, Th chip cleaning, Outside COMPOSITE	94
Th-228	9201-5, 3rd floor, Inside Delta Phase I Filter House, COMPOSITE	32
Th-228	9201-5, 3rd floor, Delta Phase I Filter House, Motor Control Room, COMPOSITE	4
Th-228	9201-5, Arc Melt, Electromelt Platform, COMPOSITE	11
Th-228	9201-5, Arc Melt, Old Machining Room, COMPOSITE	25
Th-228	9201-5, Arc Melt, COMPOSITE	122
Th-228	9201-5, Arc Melt, Knockout room COMPOSITE	23
Np-237	9201-5, 1st Floor, Main Hallway, COMPOSITE	29
Pu-238	9201-5, 2nd Floor, Mezzanine	831
Th-232	9201-5, 2nd Floor, Electronics Shop	22
Th-228	9201-5, O/S Filter Room, 3rd. Floor, Entry & Small Room, Delta Phase I COMPOSITE	7
Th-228	9201-5, Reclamation, 1st floor, Weld box, Acid Bath COMPOSITE	12
Th-228	9201-5, Scrap Reclamation Press Area COMPOSITE	55
Np-237	9201-5, 3rd Floor, COMPOSITE	39
Th-228	9202 COMPOSITE*	238
Th-228	9202, ROOF EF 622 & 633 COMPOSITE	77
Th-228	9202, Room 130 Beehive COMPOSITE	7
Th-228	9202, Room 130 Brown Dual Saw COMPOSITE	6462
Th-228	9202, Room 130 Buffalo Shear	614
Th-228	9202, Room 130 Hoods COMPOSITE	319
Th-228	9202, Room 130 Large Shear COMPOSITE	2598
Th-228	9202, Room 130 Rolling Mill	915
Th-228	9202, Room 130 Swager No.2	8888
Th-228	9202, Room 130, Extrusion Press	13802
Np-237	9202, Room 140B Glovebox	12369
Th-228	9202, Room 177 Gloveboxes COMPOSITE	12597
Np-237	9202, Room 181 COMPOSITE	4424
Th-228	9204-2E, 56, Threads COMPOSITE	2192
Th-228	9204-4, 1500 ton press COMPOSITE	14419
Th-228	9204-4, 7500 ton press COMPOSITE	1884
Th-228	9204-4, B.S., floor, Items COMPOSITE	270
Th-228	9204-4, Grinding room COMPOSITE	655
Th-228	9206 COMPOSITE	469

Th-228	9206 Room 27 COMPOSITE*	117
Th-228	9206 Room 28 COMPOSITE*	408
Pu-238	9206 Room 29 COMPOSITE*	124
Th-228	9206 Room 30 COMPOSITE*	33
Pu-238	9212 B-1 Wing, Dry Chem., COMPOSITE*	188
Th-228	9212, B-1 Wing, Primary extraction COMPOSITE	395
Th-228	9212, B-1 Wing, Primary extraction raffinate tanks COMPOSITE	272
Th-228	9212, B-1 Wing, Secondary Extraction	942
Th-228	9212, B-1 Wing, Wiped Film Evaporator	230
Pu-238	9212 C-1 Wing COMPOSITE*	381
Th-228	9212, C-1 Wing, High Capacity Evaporator	71
Th-228	9212, C-1 Wing, Intermediate Primary Extractor	890
Th-228	9212, C-1 Wing, Secondary Evaporator	919
Th-228	9212, C-1 Wing, Secondary Extractor	173
Th-228	9212, C-1 Wing, Secondary Intermediate Evaporator	174
Th-228	9212, D-1 Wing, Alligator Shear COMPOSITE	536
Pu-238	9212, D-Wing Bottle Storage COMPOSITE*	142
Pu-238	9212, D-Wing Extension COMPOSITE*	137
Th-228	9212, E-Wing Bsmt., East Hopper Room	265
Th-228	9212, E-Wing Bsmt., General Area COMPOSITE	1976
Th-228	9212, E-Wing Bsmt., West Hopper Room COMPOSITE	440
Th-228	9212, E-Wing, 150-ton Press COMPOSITE	1075
Th-228	9212, E-Wing, 75-ton Press COMPOSITE	1156
Th-228	9212, E-Wing, Bag Filter House COMPOSITE	404
Th-228	9212, E-Wing Beehive Arc Melt COMPOSITE	852
Th-228	9212 E-Wing West Casting Line COMPOSITE*	203
Th-228	9212, E-Wing, Casting Line COMPOSITE	563
Th-228	9212, E-Wing, Pack and Ship COMPOSITE	1200
Th-228	9212, E-Wing, Room 1050 COMPOSITE	427
Pu-238	9212 Room 1008 COMPOSITE*	234
Th-228	9212, Room 1010, COMPOSITE	4265
Am-241	9212, Room 1026, COMPOSITE	70
Th-228	9212, Room 1026, N Sample Cabinet COMPOSITE	332
Th-228	9212 Headhouse Room 29 COMPOSITE*	85
Pu-238	9212 Special Process Rooms 1021 and 1022/1022A COMPOSITE*	189
Th-228	9212, Special Process, COMPOSITE	1261
Th-228	9213, COMPOSITE	2883
Th-228	9215, 3rd mill COMPOSITE	3033
Np-237	9215, 3rd mill press	4413
Th-232	9215, 3rd mill press	30223
Th-228	9215, 3rd mill press	32914
Th-228	9215, 3rd mill shear COMPOSITE	6599
Np-237	9215, 52W	734
Th-228	9215/M-Wing, Chip Packing Hoods COMPOSITE	6826
Th-228	9215 Machine COMPOSITE*	282
Np-237	9720-5, 10, Item	54
Pu-238	9720-5, 17, Items, floor COMPOSITE	28

Th-228	9720-5, 20, Items	282
Th-228	9720-5, 46, Items	332
Pu-238	9720-5, 47, Item	1161
Th-228	9720-5, 48, Items COMPOSITE	83
Th-232	9720-5, 9, Items	108
Pu-239	9720-5, Conference Room, hood	1415
Th-232	9728, Dryer (Lint)	10299
Th-228	9731, Room 110/112, North Process Area	2388
Pu-238	9812 COMPOSITE*	68
Pu-238	9818 Basement COMPOSITE*	70
Th-228	9998, Development Area, G-3 lab COMPOSITE	623
Th-232	9998, H-1 Foundry, Crucible Station COMPOSITE	2421
Th-228	9998, H-1 Foundry, South Side Furnace	4288
Th-228	9998, Mold Knockout, H-1 Foundry COMPOSITE	1951
Th-228	9998, North Furnace, H-1 Foundry COMPOSITE	1800

¹"Composite" results are the sum of all individual samples in the building/area grouping.

²"U Alpha" is the total uranium activity. The Th-230 activity is included in the non-uranium alpha sum.

* Uranium to non-uranium ratios by building in 1997 study (LMES, 1997).

5.2 **IN VITRO MINIMUM DETECTABLE ACTIVITIES (MDAS), COUNTING METHODS, AND REPORTING PROTOCOLS**

5.2.1 **In Vitro Urine Analysis**

5.2.1.1 **Coverage**

Uranium enrichment activities began in the fall of 1943. A uranium urinalysis program based on fluorometry was used in conjunction with medical examinations to monitor for kidney damage from exposure to soluble uranium compounds (Sterner and Riley, 1946); the earliest fluorometry data was dated in 1948. In 1950, workers in production areas were placed on a uranium fluorometry urinalysis program for the purpose of estimating internal exposure. The program was expanded to include certain maintenance workers in 1954 (McLendon, 1960). Currently, workers with a potential for internal exposures in excess of 100 millirem per year (mrem y^{-1}) (committed effective dose equivalent {CEDE}) are required to participate in the bioassay program (BWXT Y-12, 2003a, p. 23). Until September 1989, routine urinalysis focused on EU, DU, normal uranium (NU, also known as natural uranium), tritium, and plutonium. Beginning in October 1989, uranium results were no longer classified as EU, DU, or NU. Instead, they were reported as isotopic results based on alpha spectrometry analysis. Analyses for other radionuclides were performed on an as needed basis.

5.2.1.2 **Sample Collection**

Sample Volumes

For most of the plant's history, the primary urine collection method was a spot "Monday Morning" sample submitted before entering the work area. That is, routine samples were submitted after a minimum of a 48-hour (h) absence from the work area. [In the July 1—December 31, 1951, Health Physics Progress Report (Y-858, 1952, p. 25), it was stated that "Friday evening" samples would be discontinued in favor of "Monday morning" samples.] The fraction of the daily void volume was estimated on the basis of the time between the sample voiding and the previous voiding. This "*rate method*" of estimating daily void volumes was used explicitly in the calculation of the daily radionuclide

excretion. For example, the following formula was given for calculating EU excretion by electrodeposition and gross alpha counting (Y-1401, 1965, p. 25):

$$\text{dpm/day} = 8 \times (\text{AP}/\text{Eff}) \times (\text{Vol}/\text{TI}) \quad (3.1)$$

where:

- AP = Average number of counts in 30 minutes on a plate (disk)
- Eff = Uranium recovery efficiency (%)
- Vol = Volume of total void in milliliter (mL)
- TI = Time between sample void and previous void (h)
- 8 = A constant incorporating time, count and volume constants, including the 20 mL electroplating aliquot volume.

(This equation is taken directly from the reference as shown. There is no information on what the constant includes or what the daily excretion was.) The term (Vol/TI) also appears in the corresponding equation for fluorometric determinations of NU and DU (Y-1401, 1965, p. 27). The use of the rate method to estimate daily urinary excretion (and hence, radionuclide elimination) contributed to the uncertainty associated with any given measurement and the corresponding detection level, as discussed below.

Since 1989, routine samples were collected over a 24-h period typically while the employee is on a scheduled break from the workplace. Many workers elect to submit a “simulated 24-h sample.” This sample is obtained by collecting the last void in the evening (before retiring), any urine excreted during the night, and the first void the following morning. This procedure is repeated for two consecutive nights (BWXT Y-12, 2003a, p.70).

Routine Urine Sample Frequency

Urine samples were collected monthly in 1950. Weekly collection for some employees began in 1951. By 1963, Health Physics personnel were basing the frequency of participation in the urinalysis program for each department on the most recent urinalysis results of that department. The 1963 edition of the Y-12 Radiation Safety Manual (Y-KB-29, 1963) stated that the frequency of participation schedules was reviewed monthly and adjusted semi-annually to meet the following criterion: “Sample at the frequency necessary to assure, with at least 95 percent confidence, that 95 percent of the individuals in a department have a quarterly average below the plant action limit.” This criterion was used within the limitations of a maximum frequency of once per week and a minimum frequency of once per quarter. Since the late 1980’s, most personnel were on a quarterly frequency.

The current practice is to schedule samples based on the RWP usage, and not on the basis of a default frequency. Only if an RWP was used will a person be scheduled for a sample. If the RWP specified a fecal sample, urine/fecal paired set of samples will be scheduled approximately 60 days post entry date. If the RWP specified urine only, a urine sample will be scheduled approximately 90 days post entry date.

5.2.1.3 Minimum Detectable Activities

Normal and Depleted Uranium in Urine by Fluorometry (1945—1989) and Enriched Uranium (1948—1950)

In fluorometry the visible radiation emitted from a doped, sodium fluoride bead illuminated by ultraviolet radiation is measured. Poisson counting statistics used in nuclear particle counting do not apply to this procedure. Historically, the detection limits were determined by testing the performance

of a particular configuration against standards of known content. Early Health Physics Progress Reports give the minimum detectable limit as 5 parts per billion (ppb), which is $7 \mu\text{g d}^{-1}$ for a nominal 1.4 L d^{-1} urine excretion rate (Y-339, 1949; Y-780, 1950). In 1952, the instrument sensitivity and sample volumes indicate a minimum detectable limit of $7 \mu\text{g d}^{-1}$ for 1.4 L d^{-1} of urine excreted (Y-940, 1953, p. 28).

The activity was calculated from the fluorometric mass reading for NU using a specific activity of $1.55 \text{ dpm } \mu\text{g}^{-1}$. For EU (93 weight % U-235), a specific activity of $150 \text{ dpm } \mu\text{g}^{-1}$ was used. The corresponding limits of detection are 11 dpm d^{-1} for NU and 1100 dpm d^{-1} for EU (93 weight % U-235). At times, procedures called for the direct conversion of fluorometer current into units of dpm, implicitly omitting the mass calculation. Whether the results were expressed in terms of mass or activity, the method remained essentially stable until replaced by long-duration alpha spectroscopy in 1989. Thus, from 1945 through 1989, the minimum detectable activity (MDA) is $7 \mu\text{g d}^{-1}$ or 11 dpm d^{-1} for NU. From 1948 to 1950, the MDA is $7 \mu\text{g d}^{-1}$ or 1100 dpm d^{-1} for EU (93 weight % U-235). Additional variation results from individual urine excretion volumes. Given the limitations of the rate method of estimating daily urine volumes, uncertainty in the excretion volume is expected to contribute significantly to the uncertainty associated with the detection limit of a single measurement.

Enriched Uranium in Urine by Gross Alpha Counting (1950—9/1989)

Determination of the MDAs for this method is complicated by the standard counting method used. From the mid 1950s to 1989, each “sample” consisted of two silver disks, each containing uranium electroplated from separate 20 mL raw urine aliquots drawn from the container submitted by the worker. Each disk was counted twice (on two different proportional counters), 30 min per count. If the two results from a single disk did not agree within tabulated limits, a third count was made (Y-P65-313001, 1966, p.6) and the two most concordant counts were used. If the average results of the two disks from the same sample did not agree within specified limits, then two more plates were prepared, volume permitting (Y-P65-313001, 1966, p.6). The expression of potentially censored data of this sort in terms of formal detection limits is not straightforward, and no detailed analysis of the statistics of this process has yet been located. However, an approximate indicator of the detection limit can be determined from published values, if issues regarding statistical independence are set aside.

The background count rate for the proportional counters was reported as 0.12 counts per minute (cpm) in 1963 (Y-KB-29, p.38). Counters were not used if the background was greater than 5 counts in 30 min (Hamrick, 1958, p. 7). For a disk counted for 120 min^1 , assuming a well-known background, and $\alpha = 0.05$, L_D , the detection level, (Currie, 1968) is 0.13 cpm. For 1400 mL d^{-1} urine output, a nominal 0.5 cpm/dpm counting efficiency, and an average uranium recovery of 40% as reported by Patterson (1958)), $L_D = 46 \text{ dpm per day (dpm d}^{-1})$. With an increase in average uranium recovery to 73% (Y-1401, 1965 p. 33), L_D is reduced to 25 dpm d^{-1} . In reviewing historical gross alpha urinalysis data, Barber and Forest (1995) report a “decision level” of approximately 20 dpm d^{-1} . Average uranium recoveries between 1958 and 1965 have not been identified, nor is it clear that the statistical convention used by Barber and Forest was identical to that used here. It is provisionally assumed that prior to 1965, $L_D = 46 \text{ dpm d}^{-1}$, and afterwards, $L_D = 25 \text{ dpm d}^{-1}$. However, given the limitations of the rate method of estimating daily urine volumes, uncertainty in the excretion volume is expected to add substantially to the uncertainty associated with the detection limit of a single measurement. Since recoveries were based on batch, rather than individual measurements, uncertainties in recovery would also add to the uncertainty of the detection limit of a single measurement.

¹ Actually, two disks were counted for 60 min each.

Uranium in Urine by Alpha Spectrometry (10/1989—Present)

The present alpha spectrometry system employs large urine aliquots, chemical separation of uranium, sample-specific tracers to determine recovery, and long counting times on low background detectors. The resulting L_D is approximately 0.15 dpm d⁻¹ (BWXT Y-12, 2002, p.15). The L_D varies with the uranium recovery of a particular sample, and is reported with the sample result.

Tritium in Urine by Gas Counting

Throughout most of the plant's history (earliest recorded data 1957), tritium was counted by reacting urine with calcium to evolve hydrogen gas, which was counted in an ion chamber. The MDA for this method has not been identified to date. However, the MDA was almost certainly far below the plant action level of $0.25 \mu\text{Ci mL}^{-1}$ ($8 \times 10^8 \text{ dpm d}^{-1}$) (Y-1186, 1957, p. 13). Results are reported in $\mu\text{Ci mL}^{-1}$.

Tritium in Urine by Liquid Scintillation

Currently, 5 mL of middle distillate from a urine sample is placed in liquid scintillation fluid for counting. Assuming typical method efficiencies, the MDA is 2000 dpm d⁻¹.

Plutonium in Urine by Gross Alpha Counting

The plutonium in urine procedure used prior to 1988 involved chemical separation from a 24-h void or a simulated 24-h sample prior to gross alpha counting. The detection limit has not yet been identified, but was certainly far below the corresponding limit for uranium in urine, which was based on 20 mL aliquots. If the counting method were identical (same count time – 60 min), chemical recovery (40 to 73%), and counting efficiency (0.5 cpm/dpm) to the early uranium counting method, the MDA for a 24-h plutonium sample (less 40 ml) would range from about 0.7 to 1.3 dpm per sample.

Plutonium in Urine by Alpha Spectrometry (1988—present)

The *a priori* L_D for both ^{238}Pu and for $^{239,240}\text{Pu}$ is 0.025 dpm per sample, or 0.025 dpm d^{-1} , assuming a full 24-hour void volume. Actual sample-specific MDAs are included in the analysis reports.

Other Actinides in Urine and Feces

The current analytical laboratory L_D values, since approximately 1988, are tabulated below, in units of dpm per sample (BWXT Y-12, 2003a, p.107).

Radionuclide	L_D (dpm/sample)
^{241}Am	0.05
^{228}Th	0.15
^{232}Th	0.07
^{237}Np	0.1

Uranium in Feces

Fecal sampling was started in 1998 when the presence of a less soluble component was identified as a result of the stand down. In 2002, 700 Y-12 workers participated in the fecal sampling program, out of 2800 participants in the bioassay program (BWXT Y-12, 2002, p. 7). Participation in the fecal sampling program is based on the potential for significant exposures to insoluble uranium, i.e., workers who are expected to have an internal exposure to Type S uranium and there is a potential to exceed 100 mrem (BWXT Y-12, 2003a, p.80). At present, the analytical chemistry division cites the value of 0.15 dpm per sample as a representative MDA for fecal analysis (BWXT Y-12, 2002, p. 15). Samples usually consist of a single voiding. The results in the claim files are assumed to be in units of activity per sample unless otherwise indicated (BWXT Y-12, 2003c, p. 7).

5.2.2 In Vitro Methods for Individual Radionuclides

The following section discusses the in vitro methods for specific radionuclides in urine. (Analysis of fecal samples is discussed in Section 5.2.3.)

5.2.2.1 In Vitro Bioassay for Uranium

Overview

The *in vitro* bioassay program for uranium at Y-12 can be divided into four eras. First, from 1943 to 1947, limited monitoring was performed for which the data cannot be retrieved. Second, from 1948 to 1950, fluorometric analyses of urine and blood were conducted as part of general medical surveillance to prevent kidney damage from exposure to soluble uranium compounds. Third, from 1950 to 1989, collection of routine samples based on uranium exposure potential was initiated. During this third phase, fluorometric analyses were performed on samples submitted by workers in NU and DU areas. Electrodeposition of uranium, followed by gross alpha counting, was used for samples submitted by workers in EU areas. The primary goal for the EU analysis was to control lung doses from insoluble compounds under the assumption that such materials cleared from the lung with a 120-d half life. During this lengthy era, there were modifications in procedures, but the basic approach remained the same. Fecal sampling was used for some follow-up investigations. Fourth, after October 1989, routine 24-h urine samples were collected. The uranium was extracted by chemical separation and ion exchange, and the extract was counted by alpha spectroscopy for an extended period (1000 min per sample). Since 2000, fecal samples are submitted routinely for individuals working with largely insoluble forms of uranium.

Uranium Analysis by Fluorometry: (1948—1989)

Uranium processing began in the fall of 1943. Y-12 technical reports describe the use of fluorometry for uranium detection as early as 1944 (Van Wazer and Reiss, 1944). By 1946, fluorometric analyses for uranium in urine and blood were in general use to supplement clinical surveillance for soluble uranium exposures (Stern and Riley, 1946). Fluorometric uranium urinalysis data from the Tennessee Eastman Corporation era (1943—1947) were discussed in a 1981 mortality study of plant personnel (Polednak and Frome, 1981).

Early in the Union Carbide management of Y-12, the fluorometric method was reassessed. Extraction methods were used starting in 1949 and 1950 (Y-365, 1949, p. 7; Y-780, 1950, p. 14). By the first half of 1952, a technique involving small (0.2 mL) aliquots of raw urine had been put in place (Y-940, 1953, p.28). However, after this change in technique was instituted, the urinary uranium concentrations increased greatly, as shown in Table 5-6 on p. 27. This raises the concern that the technique used prior to May of 1952 may have underestimated the urinary uranium concentrations. (See Section 5.4.2 regarding the discussion of missed doses.)

Fluorometric analysis continued to be used for NU and DU urinalysis until 1989, when alpha spectroscopy began to be used for all uranium urinalysis. Fluorometry yields results in mass units (e.g., $\mu\text{g L}^{-1}$) but results were often converted to dpm d^{-1} with the assumed specific activity of $1.55 \text{ dpm } \mu\text{g}^{-1}$ for NU. The fluorometric technique had an industry standard sensitivity of about 5 ppb, or $5 \mu\text{g L}^{-1}$ (e.g., Y-339, 1949, p. 7).

Uranium Analysis by Electrodeposition and Alpha Counting: (1950—1989)

The primary objective of this procedure was to prevent EU lung burdens in excess of the prevailing limit, $0.017 \mu\text{Ci}$, corresponding to an ICRP 2 calculated lung dose of 15 rem y^{-1} ($\text{QF} = 10$). Samples from workers in EU areas were analyzed by electrodeposition of uranium onto silver discs, which were then counted for gross alpha activity in a proportional counter. Development of the gross alpha

counting method was reported to be underway in early 1949 (Y-365, 1949, p. 7). At first, samples were subjected to an acid digestion step prior to electrodeposition. In 1951, methodological problems leading to underestimates of a factor of two were noted and corrected (Y-858, 1952, p. 53). The information available was not sufficient to determine if the records were corrected. In the mid 1950's, the acid digestion step was discontinued altogether due to reported contamination problems, as well as for logistical considerations (Patterson, 1958, p. 34).

From the mid 1950's forward, two 20-mL aliquots of raw urine from each sample were placed in electrodeposition cells, producing two discs per sample. Each disc was counted once on one proportional counter for 30 min and a second time on a different counter, and the resulting counts were averaged. The background count rate was reported as 0.12 cpm in 1963 (Y-KB-29, 1963, p.38). The expected net count rate for a person excreting at the action level of 70 dpm d⁻¹ was 0.2 cpm, given a urine output of 1.4 L d⁻¹, a nominal 0.5 dpm/cpm counting efficiency, and a uranium recovery of 40%. Because each disk was counted twice for 30 min each, the expected net count for the two counts on each disk under these conditions was 12 counts, over a background of 7.2 counts.

Recoveries were estimated on the basis of spiked samples that accompanied each counting run. The typical "spike" contained relatively little activity. Recoveries from raw urine tended to be around 40% in the late 1950s, (Patterson, 1958), but improved over time. In 1965, the average recovery for electroplating was 73% (Y-1401 1965, p.33). Method limitations are discussed in a number of reports, including Johnson et al. (1959) and Y-B2-125 (1959). Recoveries were found to vary with pH, drying procedure, and uranium content, with recoveries decreasing for uranium concentrations below the plant action level. The precision of any one sample was acknowledged to be relatively low:

Further, realizing the over-all lack of precision in any one sample result, we normally recommend restriction only on the basis of the 13-week or quarterly cumulative internal dose as indicated by from 2 to 13 samples. (Patterson 1958, p. 57)

The precision of an individual result in this type of procedure is poor and for this reason little significance is attached to individual results, particularly those below the "Plant Action Limit (PAL)" level. (Y-P65-313001, 1966).

These limitations notwithstanding, the salient point is that, when *in vivo* counting became routine in the early 1960s, very few additional workers were found to require restriction from uranium areas (e.g., Y-KB-33, 1963, p.6). The *in vivo* monitoring frequency was determined by urinalysis results and ranged from monthly to once/18 months (Y-KB-33 1963, p.1). Almost all of the restrictions that did take place would have occurred on the basis of the urinalysis program alone. In those cases in which restriction was based on *in vivo* analysis alone, lung retention times were often observed to be considerably longer than the assumed 120-d lung half-life on which the urinalysis program was based. In other words, the problem was largely due to the inapplicability of the lung model for some materials and/or individuals, rather than a failure of the urinalysis program *per se*.

Uranium-233 by Gross Alpha Counting

In early 1962, Y-12 undertook the fabrication of metallic ²³³U from a nitrate solution (U-233 uranyl nitrate solution) in building 9205 (West and Roberts, 1962). In recognition of the high specific activity of this material, and the potential for gamma radiation from the daughter products of contaminant ²³²U, operations were doubly contained. Pilot runs were made with less hazardous materials to test the protective measures. As part of these precautions, eight workers submitted urine samples for analysis by electrodeposition and gross alpha counting as described above, with the same recoveries and detection efficiencies. This project lasted 3 months and was completed in 1962. Ratios of U-233 to U-232 varied with operation and time. Ratios for various exposure scenarios are provided in the

following table. In addition to absorption Types M and S, exposure to Type F material is also considered likely in this process. U-232 has the larger dose conversion factor.

Process	U-233:U-232 Activity Ratio
Reduction of UF ₄	1.3
Casting of metal-crucible skull	15-32
Machining final uranium component	160

Due to uncertainty in the process, claimant favorable assumptions should be made regarding solubility and uranium activity ratios.

Uranium Analysis by Alpha Spectrometry: (1989—present)

Up to 2 L of urine, along with a ²³²U tracer, are treated with nitric acid (HNO₃) and hydrogen peroxide (H₂O₂). The sample is wet ashed, and the uranium is co-precipitated with calcium oxalate (CaC₂O₄). After dissolving the precipitate in hydrochloric acid (HCl), the uranium is further separated by ion exchange chromatography. The uranium is eluted from the column with a solution of dilute HCl to which titanous chloride (TiCl₃) has been added to reduce actinides that may be in an elevated oxidation state. The final fraction of the eluate is first treated with ascorbic acid to reduce the presence of any ferric iron and next with hydrofluoric acid (HF). Then the uranium isotopes are co-precipitated on neodymium fluoride (NdF₃). The NdF₃ is caught on a 0.1 millimeter (mm) filter, rinsed, dried, and then mounted on a planchet for alpha spectrometry (BWXT Y-12, 2003a). Typical recoveries are about 85%. Samples are typically counted for 16 h on a passivated implanted planar silicon (PIPS) detector.

5.2.2.2 In Vitro Bioassay for Plutonium

Plutonium in Urine by Gross Alpha Counting

In 1957, certain workers in the Special Testing Department were tested for plutonium intake by urinalysis. Because large urine volumes were required, these employees submitted 24-h samples through the use of take-home kits. The sampling frequency was monthly (Y-1186, 1957, p.38). After two 20-mL aliquots were removed for uranium analysis by electrodeposition, the balance of the sample was treated by chemical separation, precipitation, and evaporation onto a stainless steel planchet for alpha counting. Health physics personnel converted the reported activity rate (dpm per 24 h void) to mrem d⁻¹ to the critical organ.

Uranium, Plutonium, and Americium by Alpha Spectrometry (10/1989 – Present)

Up to 2 L of urine, along with ²³²U, ²⁴²Pu, and ²⁴³Am tracers, are treated with HNO₃ and H₂O₂. The sample is wet ashed, and the uranium, plutonium, and americium are co-precipitated with CaC₂O₄. After dissolving the precipitate in HCl, uranium, plutonium, and americium are further separated by the use of two ion exchange columns and an additional oxalate precipitation. The uranium, plutonium, and americium in the final fractions are co-precipitated with NdF₃. The NdF₃ is caught on a 0.1 mm filter, rinsed, dried, and then mounted on a planchet for alpha spectrometry (BWXT Y-12, 2003a). The same procedure is used for isotopic thorium.

5.2.2.3 *In Vitro* Bioassay for Tritium

As of 1957, personnel engaged in processing materials with a potential for tritium contamination submitted three urine samples per month. Samples were submitted at the same stations used for uranium and plutonium samples. The urine samples were reacted to evolve hydrogen gas, which was then counted for beta activity in an ionization chamber. Results were reported in terms of $\mu\text{Ci mL}^{-1}$. (Y-1186, 1957, pp.38-39)

Currently, tritium is measured by liquid scintillation counting. An aliquot of the urine sample is distilled in a heating mantle, and the middle fraction of the distillate is collected. A 5-mL portion of the collected distillate is mixed with liquid scintillation cocktail. The beta activity of the tritium is then measured by liquid scintillation counting (BWXT Y-12, 2003a). Exposure should be assumed to be due to HTO.

5.2.2.4 *In Vitro* Bioassay for Other Radionuclides

The following alpha spectrometry procedures described in Section 3.2 of BWXT Y-12 (2003a) have been used since 1989.

Neptunium-237 in Urine by Alpha Spectrometry

In this procedure, ^{239}Np is used as an internal yield monitor. Americium-243, which is in equilibrium with its daughter, ^{239}Np , is added during the sample preparation process. The separation time of ^{239}Np from its parent, ^{243}Am , is recorded (so that the count can be decay corrected), and the beta activity is measured to calculate the radiochemical yield. The tracer is added to the urine sample (or fecal material) along with concentrated HNO_3 and H_2O_2 , and the sample is wet ashed. Neptunium is co-precipitated with CaC_2O_4 and mixed hydroxides. After dissolving the precipitate in concentrated HNO_3 and ashing in a muffle furnace, the oxidation state of neptunium is adjusted to the tetravalent state by using ferrous sulfamate as a reducing agent. Neptunium is separated from other actinides by the anion exchange method. Neptunium is eluted with 0.5 M HCl containing TiCl_3 . The eluate is collected, and neptunium is co-precipitated on NdF_3 and filtered on 0.1 mm filter paper. The NdF_3 is rinsed, dried, and mounted on a planchet for beta counting and alpha spectrometry.

Isotopic Thorium (Th-228, Th-230, and Th-232) by Alpha Spectrometry

Up to 2 L of urine along with a ^{229}Th tracer is treated with HNO_3 and H_2O_2 . The sample is wet ashed, and the thorium is co-precipitated with CaC_2O_4 . After dissolving the precipitate in HNO_3 and mixing with methanol, the thorium is further separated by ion exchange chromatography. The thorium is eluted from the column with 8 M HCl. Hydrofluoric acid (HF) is added, and the thorium isotopes are co-precipitated with NdF_3 . The NdF_3 is caught on a 0.1 mm filter, rinsed, dried, and then mounted on a planchet for alpha spectrometry. The same procedures are used for uranium, plutonium, and americium determinations. This method accommodates substantial quantities of common interferences, such as iron, aluminum, titanium, zirconium, and uranium. However, plutonium can present some interference. If plutonium is expected, an alternate method should be chosen (BWXT Y-12, 2003a).

5.2.3 Fecal Sample Analysis

Fecal samples have long been used in follow-up investigations when urinalysis or *in vivo* measurements indicate the likelihood of a substantial intake. West and Scott (1966) describe such an investigation in the 1960s, in which fecal, urine, and *in vivo* measurements were used jointly to investigate clearance. Fecal sampling was discontinued several times in the past, but was reinstated in 1998 due to changes in workplace exposure conditions. At first, fecal sampling was used for a limited number of workers in Building 9212, but was expanded to include Building 9215 in 1999 and to

other areas of the plant in 2000 and 2001 (BWXT Y-12, 2002, p. 6).

If a work area has been determined to have predominantly insoluble airborne uranium, it is current practice for workers with a moderate to high exposure potential (CEDE > 100 mrem) to submit both urine and fecal samples. The relative elimination by urinary and fecal pathways is used to determine the solubility mixtures for each individual. For combined urine and fecal monitoring, the routine sampling interval is 53 d (BWXT Y-12, 2002, p. 14). Twenty-four hour fecal collections are preferred. When 24-hour fecal samples are not available, the results can be normalized using the ratio of the Reference Man excretion rate of 135 g d⁻¹ to the mass of the submitted sample. The Y-12 practice was to routinely normalize results to reference man. These normalized results or actual individual daily excretion results may be used to reconstruct dose.

5.2.4 Interferences

As investigation levels decreased over time, the contribution of natural background to worker uranium excretion assumed greater importance. Since 1989, Y-12 has corrected the measured uranium excretion values for background uranium interference when performing dose assessments. However, the uranium results that are reported in response to NIOSH requests for dose records have not been corrected for dietary uranium.

Y-12 used the following methods to correct sample results reported after 1989 for dietary levels of uranium. The range of background uranium values expected to be observed in urine samples is 0.14 dpm – 0.57 dpm and the range of background uranium values expected to be observed in the fecal samples is 2.32 dpm – 2.75 dpm (BWXT Y-12, 2003a, p. 75). Therefore, at Y-12, the reference point is established at the midpoint of the quoted ranges of values for urine (0.35 dpm d⁻¹) and fecal samples (2.5 dpm/sample). The ratios between ²³⁴U and ²³⁸U can also provide information to help characterize the sample. Natural background uranium typically has a ²³⁴U to ²³⁸U ratio of approximately 1:1. Enriched uranium also has a characteristic ratio, typically greater than 3:1. Depleted uranium, on the other hand, typically has a characteristic ratio of less than 0.9:1.

Based on the above information, the background uranium corrections used at Y-12 are as follows:

For bioassay sample results less than the detection limit for the analytical technique, no action is required. Note, that corrections for samples from the early days when MDAs were significantly larger are not required.

To adjust results for background, the ²³⁴U to ²³⁸U ratios are reviewed for each sample to determine if the sample is consistent with EU or DU. Conservative ratios are applied in this determination. All samples with ²³⁴U to ²³⁸U ratios greater than 1:1 are considered to be consistent with EU and all samples with ²³⁴U to ²³⁸U ratios less than 1:1 are considered to be consistent with DU.

The background contribution is subtracted from EU samples as follows:

- a) Assume that the ratio of ²³⁴U to ²³⁸U is 1:1 for dietary uranium.
- b) Determine if the ²³⁸U result exceeds its corresponding critical level value.
- c) If the ²³⁸U result is greater than the critical level, then subtract two times that ²³⁸U result from the total uranium result.
- d) If the ²³⁸U result is less than its corresponding critical level value, do not subtract a dietary component.

For DU samples, e.g., those with ^{234}U to ^{238}U ratio less than 1:1, the following steps are used to determine the dietary correction for DU samples:

- a) The following equations are solved to determine an equation for the occupational ^{238}U content and the occupational ^{234}U content in a DU sample:

$$\frac{^{234}\text{U}_{\text{diet}}}{^{238}\text{U}_{\text{diet}}} = 1 \quad (3.2)$$

$$\frac{^{234}\text{U}_{\text{occ}}}{^{238}\text{U}_{\text{occ}}} = 0.2 \quad (3.3)$$

$$^{238}\text{U}_{\text{actual}} = ^{238}\text{U}_{\text{diet}} + ^{238}\text{U}_{\text{occ}} \quad (3.4)$$

$$^{234}\text{U}_{\text{actual}} = ^{234}\text{U}_{\text{diet}} + ^{234}\text{U}_{\text{occ}} \quad (3.5)$$

- b) Using Equation 3.5, and substituting the $^{238}\text{U}_{\text{diet}}$ for the $^{234}\text{U}_{\text{diet}}$ based on Equation 3.2 and substituting 0.2 ($^{238}\text{U}_{\text{occ}}$) based on Equation 3.3 for the $^{234}\text{U}_{\text{occ}}$, the following relationship results:

$$^{234}\text{U}_{\text{act}} = ^{238}\text{U}_{\text{diet}} + 0.2(^{238}\text{U}_{\text{occ}}) \quad (3.6)$$

Since the $^{238}\text{U}_{\text{diet}}$ can be replaced with the following relationship derived from Equation 3.4: $^{238}\text{U}_{\text{actual}} - ^{238}\text{U}_{\text{occ}}$, Equation 3.5 can be solved to determine the occupational quantity of ^{238}U . The resulting equation for determining the occupational quantity of ^{238}U is:

$$\frac{^{234}\text{U}_{\text{act}} - ^{238}\text{U}_{\text{act}}}{-0.8} = ^{238}\text{U}_{\text{occ}} \quad (3.7)$$

- c) Once the occupational ^{238}U quantity is known, the occupational quantity of ^{234}U can be derived using Equation 3.3:

$$^{234}\text{U}_{\text{occ}} = 0.2(^{238}\text{U}_{\text{occ}}) \quad (3.8)$$

- d) Ensure that both the ^{234}U and ^{238}U results exceed their corresponding critical level values. If both of these results are not greater than their corresponding critical level values, then a background determination cannot be made. In this case, it is assumed that all of the activity observed in the total uranium sample result will be used to determine occupational dose.
- e) Use either Equation 3.7 or 3.8 to determine the occupational ^{238}U component or the occupational ^{234}U component, respectively. For ease of reference, the ^{238}U component was chosen for the remaining steps.
- f) Subtract the occupational component of the sample from the corresponding observed component to determine the dietary contributor:

$$^{238}\text{U}_{\text{act}} - ^{238}\text{U}_{\text{occ}} = ^{238}\text{U}_{\text{diet}} \quad (3.9)$$

- g) Once the dietary component has been calculated, subtract twice this dietary value from the total uranium result to remove the background:

$$U_{tot} - 2(^{238}\text{U}_{diet}) = U_{totalocc} \quad (3.10)$$

5.3 ***IN VIVO* MDAS, COUNTING METHODS, AND REPORTING PRACTICES**

5.3.1 **Whole Body Counting**

Whole body counting was not routinely practiced at Y-12. The primary *in vivo* detection method was chest counting, as described below.

5.3.2 **Chest Counting**

The Y-12 *in vivo* chest counting facility was developed in the late 1950's and was put into routine use in 1961. The original facility is described in articles by R. E. Cofield (Cofield 1959; Cofield 1960). A 9" x 4" sodium iodide (thallium doped) [NaI(Tl)] crystal was placed over the chest of a subject reclining in a cot in a shielded room. Subjects showered, shampooed, and changed into a clean garment prior to counting. They were then surveyed for any remaining surface contamination before entering the counting room. The normal counting time was 20 min.

Incremental improvements in the system were made over time. A second 9" x 4" NaI(Tl) crystal was placed under the subject's back in 1963 or 1964. It was found that this position provided greater counting efficiency than the position over the chest. A second advantage was that skin contamination seldom occurred on the back. The ratio of the count rates from the two crystals could be used to identify surface contamination missed by the survey meter. In 1965, two 5" NaI(Tl) crystals were added, positioned at the sides under the arms. In 1985, initial development of a high purity germanium (HPGe) counting system was begun. The new system began routine operation in a new counting room in June 1992.

5.3.2.1 **Chest Counting for Uranium**

With the NaI scintillation system used for much of the plant's history, the amount of uranium deposited in the lungs was inferred from the worker's spectrum by use of a prediction equation. This included the subject's weight and constants derived from the spectra of control subjects. Compton scatter from natural ^{40}K and fallout ^{137}Cs in the body was a particularly important contributor to background in the ^{235}U region of interest around 186 kiloelectronvolt (keV) and the ^{238}U region of interest around 93 keV. The ^{137}Cs contribution to the regions of interest changed over time as fallout levels varied, requiring frequent updates to the control subject spectrum library.

Measured lung burdens were originally expressed in μg of ^{235}U for EU and mg of ^{238}U for NU or DU. The original limits of detection for ^{235}U and ^{238}U were reported as 130 μg and 13.5 mg, respectively (Cofield, 1959). Improvements in hardware and data reduction procedures led to lower detection limits, as indicated in Tables 5-4 and 5-5 below. Limits are given both in mass units and in terms of activity (nanocurie [nCi] and dpm), with the published value in regular type and the converted value in italics. When known, the reporting convention used to define the detection limit is indicated in the tables. The alternative definitions listed by King and Barclay (1983) illustrate the importance of this factor.

Due to the uncertainty regarding actual methods for determining the lung counting detection limits, for dose reconstruction purposes and based on review of the Y-12 data, the U-235 and U-238 lung count

detection thresholds are assumed to be 130 µg and 13.5 mg, respectively, through 1990. Beginning in 1991, the critical level or MDA is supplied with the individual results.

No description of the conversion count rate to activity to mass has been found. The analyte reported was based on the area in which the employee worked. Individuals working in NU or DU areas had results reported as U-238, and workers in enriched areas had results reported as U-235. Claimant favorable assumptions should be based on conversions of 93% enrichment for U-235 and natural isotopic abundances for U-238.

5.3.2.2 Chest Counting for Other Actinides

Thorium

The *in vivo* lung count was the only monitoring technique for monitoring thorium exposure in the body during the plant's first decades. Thorium lung activity was inferred from ^{228}Ac and/or ^{212}Pb lung activity. Thorium lung counting was conducted from 1958–1984, with routine lung counts starting in 1961 scheduled at approximately six-month intervals.

Table 5-4. Reported detection limits for ^{235}U .

Year	Detectors	^{235}U (µg)	^{235}U (nCi)	^{235}U (dpm)	Reporting convention	Reference
1959	One 9" NaI	130	0.28	620		Cofield 1959
1963	Two 9" NaI	96	0.21	470		Scott and West 1967
1965	Two 9", 2 5" NaI	72	0.16	360		Scott and West 1967
1975	Two 9", 2 5" NaI	70	0.15	330		Scott and West 1975
1983	Two 9", 2 5" NaI	62	0.13	290	$1.96 \sigma_{\text{BKG}}$	King and Barclay 1983
1983	Two 9", 2 5" NaI	68.5	0.15	330	L_C^a	King and Barclay 1983
1983	Two 9", 2 5" NaI	137	0.30	670	L_D^a	King and Barclay 1983
1990	Two 9", 2 5" NaI	70	0.15	330	b	Barber and Forest 1995
6/1992	HPGe	46	0.10	220	c	BWXT Y-12 2003a

a. As defined by Currie, 1968, with $\alpha = \beta = 0.05$, paired observations.

b. Type I error = 5%.

c. Individual MDA or critical level supplied with analytical results.

Table 5-5. Reported detection limits for ^{238}U .

Year	Detectors	^{238}U (mg)	^{238}U (nCi)	^{238}U (dpm)	Reporting convention	Reference
1959	One 9" NaI	13.5	4.5	10,000		Cofield 1959
1965	Two 9", 2 5" NaI	6.5	2.2	4,900		Scott and West 1967
1983	Two 9", 2 5" NaI	5	1.7	3,800	$1.96 \sigma_{\text{BKG}}$	King and Barclay 1983
1983	Two 9", 2 5" NaI	7.2	2.4	5,300	L_C^a	King and Barclay 1983
1983	Two 9", 2 5" NaI	14.4	4.8	11,000	L_D^a	King and Barclay 1983
1991	HPGe	4.5	1.5	3,300	b	BWXT Y-12 2003a

a. As defined by Currie, 1968, with $\alpha = \beta = 0.05$, paired observations.

b. Individual MDA or critical level supplied with analytical results.

The interpretation of thorium spectra is complicated by the tendency of decay chain members to become separated during operations, particularly those involving heat. The boiling point of radium is below the melting point of thorium. The ^{228}Ac gamma radiation in an *in vivo* count could be an indication of a ^{228}Ra intake or an intake of the entire ^{232}Th chain. The 239 keV gamma line from ^{212}Pb , a daughter radionuclide of ^{228}Th , is a more reliable indicator of ^{232}Th , as the thorium isotopes would remain together during processing. However, if the ^{228}Ra is lost, ^{212}Pb will decrease with the 1.9 y half life of its parent, ^{228}Th , moderated by the ingrowth of new ^{228}Ra with its 5.7 y half-life. This complicated disequilibrium pattern operates both before and after chain members are taken into the

body. Some knowledge of the likely state of equilibrium is therefore necessary in order to translate observed activities into dose.

The maximum permissible lung burden (MPLB) of ^{232}Th (corresponding to 15 rem y^{-1} to the lung) varied markedly with the ^{228}Th to ^{232}Th ratio. For full equilibrium (ratio = 1) the MPLB was calculated to be 2.9 nCi of ^{232}Th . For a ^{228}Th : ^{232}Th ratio of 0.1, the lung burden was 12 nCi (West, 1965, p. 22). For a ratio of 0.8, an MPLB of 3.2 nCi is indicated. For this ratio, with a ^{228}Ra to ^{232}Th ratio of 0.6, an MDA of 0.2 lung burdens, or 0.6 nCi is indicated (West, 1965, p. 26). In mass units, this is 5.5 mg.

It was reported in 1965 that thorium at Y-12 was processed less than one year after purification by the supplier, and consequently had only about 10% as much ^{228}Ra as ^{224}Ra (West, 1965, p. 18). This means that the maximum dose conversion factor per mg of ^{232}Th would be less than that for ^{232}Th in full equilibrium with its progeny. However, unless specific information is available with the claimants' data, the dose reconstructor will have to make claimant-favorable assumptions. The thorium sensitivity varied due to the dependence of this technique on the ratios of Th-232 and Ra-228 to the daughter radionuclide being measured. Data interpretation was based on a careful evaluation of the work histories. In the absence of sufficient data to determine Th-228:Th-232 ratios and state of equilibrium, the claimant favorable assumption is to assume full equilibrium.

Neptunium

At the time the *in vivo* system was put into routine service in 1961, the reported detection limit for ^{237}Np without daughter radiation was 2.7 nCi (Cofield, 1961). For ^{237}Np in full equilibrium with ^{233}Pa , the detection limit was reported as 0.255 nCi (Cofield, 1961).

5.3.2.3 Chest Counting for Other Radionuclides

At the time the *in vivo* system was put into routine service in 1961, the reported detection limit for ^{60}Co was 0.66 nCi. For ^{95}Zr in transient equilibrium with ^{95}Nb , the reported detection limit was 1 nCi. Although Bremsstrahlung counting may have been done for ^{99}Tc , no information is available regarding the sensitivity of the technique.

5.4 MISSED DOSE

There are two eras of operations at Y-12 for which sufficient monitoring information may not be available. From 1943 to 1947, limited or no monitoring was performed. No data has been found for this time period. The primary site activities for this time period were uranium enrichment by the Calutron process. From 1948 to 1950, fluorometric analyses of urine and blood were conducted as part of general medical surveillance to prevent kidney damage from exposure to soluble uranium compounds; these data cannot be retrieved at this time. Major site activities for the 1948 to 1950 period (and through 1992) included the manufacture of nuclear weapons components.

5.4.1 Data for Missed Dose Determination – 1943-1947

This section is in preparation. Please note that this era consisted primarily of enrichment activities using the Calutron process. It is anticipated that reconstruction activities will have to be based on air sampling results.

5.4.2 Data for Missed Dose Determination – 1948-1951

The primary site activities from 1948 to 1992 were associated with the manufacture of nuclear weapons components. The information in Table 5-6 is based on a review of bioassay data reported

in Health Physics Progress Reports that include data from October 1950 through June 1953. These reports presented uranium concentrations in worker's urine samples at various percentiles. The data used in this analysis are provided in Table 5-6 below.

Table 5-6. Summary of 90th percentile uranium concentrations in urine.

Date	Fluorometric Results (µg/24 h)	Gross Alpha Results (dpm/24 h)	Reference
10/1950	7	72	Y-780, 1951
11/1951	4	50	Y-780, 1951
12/1950	6	58	Y-780, 1951
01/1951	3	50	Y-836, 1951
02/1951	4	62	Y-836, 1951
03/1951	2	70	Y-836, 1951
04/1951	3	46	Y-836, 1951
05/1951	6	34	Y-836, 1951
06/1951	6	38	Y-836, 1951
07/1951	10	55	Y-858, 1952
08/1951	10	50	Y-858, 1952
09/1951	10	50	Y-858, 1952
10/1951	9	50	Y-858, 1952
11/1951	8	90	Y-858, 1952
12/1951	8	90	Y-858, 1952
01/1952	No data	83	Y-940, 1952
02/1952	No data	92	Y-940, 1952
03/1952	No data	92	Y-940, 1952
04/1952	No data	92	Y-940, 1952
05/1952	39	77	Y-940, 1952
06/1952	41	72	Y-940, 1952
07/1952	23	30	Y-1066, 1954
08/1952	25	47	Y-1066, 1954
09/1952	35	58	Y-1066, 1954
10/1952	34	92	Y-1066, 1954
11/1952	23	68	Y-1066, 1954
12/1952	48	91	Y-1066, 1954
01/1953	22	81	Y-1070, 1954
02/1953	18	70	Y-1070, 1954
03/1953	19	48	Y-1070, 1954
04/1953	19	70	Y-1070, 1954
05/1953	26	72	Y-1070, 1954
06/1953	22	90	Y-1070, 1954

Averaging the available 90th percentile values yields estimates of 17 µg/sample for samples analyzed fluorometrically and 66 dpm/sample for samples analyzed by gross alpha. However, the fluorometric data for the period from October 1950 to December 1951, appear to be much lower than the values from May 1952 through June 1953. The average concentration in the earlier period is 6.4 µg/24 h, compared to 28.1 µg/24 h for the latter period. A new technique was initiated in May 1952. No information is available to determine whether this discrepancy is related to the change in technique.

The concern is whether the urinary uranium concentrations reported in the worker's record prior to May 1952 are valid measures of the total uranium in urine. The average concentration for the latter period, i.e., 28.1 µg/24 h, could be used in lieu of the worker's measured concentration from 1948 to 1951. The dose reconstructor can use the measured concentrations from May 1952 onward or the average concentration, 28.1 µg/24 h, if no individual monitoring results are available.

If the work area or form of uranium present can not be determined, the gross alpha value should be used for determining the missed dose. Use other parameters for dose reconstruction as provided in the previous sections of this report.

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GLOSSARY

activity median aerodynamic diameter (AMAD): The diameter of a sphere having a density of 1 g/cm^3 and the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol.

acute exposure: Exposure of short duration.

background: Radiation from:

- Naturally occurring radioactive materials which have not been technologically enhanced;
- Cosmic sources;
- Global fallout as it exists in the environment (such as from the testing of nuclear explosive devices);
- Radon and its progeny in concentrations or levels existing in buildings or the environment which have not been elevated as a result of current or prior activities; and
- Consumer products containing nominal amounts of radioactive material or producing nominal amounts of radiation.

bioassay: The determination of kinds, quantities, or concentrations, and, in some cases, locations of radioactive material in the human body, whether by direct measurement or by analysis and evaluation of radioactive materials excreted or removed from the human body. Another word for radiobioassay.

chronic exposure: Exposure of long duration, delivered by fractionation or protraction.

class D, W, or Y material: ICRP 30 classifies inhaled radioactive materials as D, W, or Y (days, weeks, or years) depending on their retention time in the pulmonary region. Class D materials have a pulmonary half-time of less than 10 days; W materials, a half-time from 10 to 100 days; and Y, greater than 100 days. The times actually used for these classes of materials in ICRP 30 in the calculation of ALIs are 0.5 days, 50 days and 500 days for class D, W, and Y material, respectively. Note: A special class Y uranium has been defined as having an effective pulmonary half-life of 100 days and the systemic distribution, retention, and excretion parameters of class W uranium. (ANSI, 1995)

class (Type) Q system: an $8 \mu\text{m}$ AMAD is used as the basis for the "class Q" dosimetry system and is described as 10% class Y and 90% modified class W (the modification consisting of increasing the class W 50 day compartment to 120 days).

committed effective dose equivalent (CEDE): The sum of the committed dose equivalents to various tissues or organs in the body for fifty years post intake. CEDE is expressed in units of rem (or sievert). (IG-C1)

depleted uranium: Depleted uranium, a byproduct of the enrichment process, has a reduced abundance of ^{235}U relative to the abundance of 0.7% in natural uranium.

direct radiobioassay: The measurements of radioactive material in the human body utilizing instrumentation that detects radiation emitted from the radioactive material in the body (synonymous with *in vivo* measurement.)

dose: A general term for absorbed dose, dose equivalent, effective dose equivalent, committed dose equivalent, committed effective dose equivalent, or total effective dose equivalent as defined in this part.

enriched uranium: Uranium with an increased abundance of ^{235}U . Enriched uranium varies from greater than the natural ^{235}U abundance of 0.72% up to almost 100%. Enriched uranium may also contain an increased abundance of ^{234}U and might also contain ^{236}U and ^{233}U .

exposure: (1) The general condition of being subjected to radiation, such as by exposure to radiation from external sources or to radiation sources inside the body. In this document, exposure does not refer to the radiological physics concept of charge liberated per unit mass of air. The product of exposure time to a radioactive aerosol and the average concentration during exposure, divided by the value of the DAC for the radioactive material in question (expressed in DAC-h).

indirect radiobioassay: Measurements to determine the presence of or to estimate the amount of radioactive material in the excreta or in other biological materials removed from the body (synonymous with *in vitro* measurement.) (ANSI, 1996)

intake: The amount of radionuclide taken into the body by inhalation, absorption through the skin, injection, ingestion, or through wounds.

***in vitro* measurement:** Synonymous with indirect radiobioassay.

***in vivo* measurement:** Synonymous with direct radiobioassay.

maximum permissible body burden (MPBB): Limit for body content of a radionuclide, used from startup until 1975.

maximum permissible lung burden (MPLB): During the 1970's the occupational limit for PU was expressed in terms of a quantity of plutonium that could be present in the chest at any given time, equal to 16 μCi (0.25 μg) ^{239}Pu .

minimum detectable amount (MDA): The smallest amount (activity or mass) of an analyte in a sample that will be detected with a probability β of non-detection (Type II error) while accepting a probability of erroneously deciding that a positive (non-zero) quantity of analyte is present in an appropriate blank sample (Type I error).

minimum detectable concentration (MDC): The minimum detectable amount (MDA) expressed in units of concentration. (ANSI, 1996)

monitoring: The measurement of radiation levels, airborne radioactivity concentrations, radioactive contamination levels, quantities of radioactive material, or individual doses and the use of the results of these measurements to evaluate radiological hazards or potential and actual doses resulting from exposures to ionizing radiation.

natural uranium: The mixture of uranium as it is found in nature (i.e., 99.3% ^{238}U , 0.7% ^{235}U , and 0.0058% ^{234}U by weight). The alpha activity comes mostly, and in about equal amounts, from ^{238}U and ^{234}U . The specific activity of this mixture is $2.6 \times 10^7 \text{ Bq kg}^{-1}$ (0.7 pCi g $^{-1}$).

nephrotoxicity: poisoning of the kidney

quality factor: A modifying factor used to calculate the dose equivalent from the absorbed dose.

routine bioassay monitoring: Any bioassay measurement made on a predetermined, periodic schedule, to establish a worker's internal exposure status relative to previous periods of time.

sensitivity (detection sensitivity): The minimum amount of contaminant (or dose) than can be repeatedly measured (or calculated) by a particular analysis. See definition for minimum detectable amount (MDA).

special bioassay monitoring: Any bioassay measurement that is not required for routine bioassay, but that is required for confirmation of a suspected intake of radionuclides, or is required for follow-up evaluation of confirmed intakes. It also includes declared pregnant worker bioassay monitoring.

Type F, M, or S material: ICRP 66 (1994) classifies inhaled radioactive materials as F, M, or S (fast medium or slow) depending on their retention time in the pulmonary region. These designations are similar to the clearance classes D, W, and Y, but refer strictly to the rate of absorption from the lungs to the blood.

ATTACHMENT 5D

OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS

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5A.1 CONVENTIONS USED IN INTERNAL DOSE REPORTS

The following is adapted from Souleyrette (2003).

5A.1.1 Y-12 Urinalysis Data Before 10/89

Dept – Department number of employee at the time of monitoring

Badge/Old SSN – Employee identification number

Void Date – Date sample was voided

Program Code

- 1 = enriched uranium urinalysis by gross alpha
- 2 = natural or depleted uranium by fluorophotometry (used for enriched uranium prior to 1950)
- 6 = control sample
- 9 = (may have been used for a limited time to indicate experimental analyses methods)

Volume – volume of sample in mL

Time – Time between last void and sample void. Used to normalize urine result to 24-h voiding based on individual employee daily excretion rate. (A zero time in conjunction with a large void volume and program code 9 probably indicates an experimental measurement technique.)

Uranium Result (dpm) – The normalized urine result in dpm/d

Background Value – Background was stored for later results. The column Uranium Result (dpm) is background corrected already.

5A.1.2 Y-12 Urinalysis Data Since 10/89

Dept - Department number of employee at the time of monitoring

Badge - Employee identification number

Sample Number – laboratory sample identifier

Sample Date - Date sample was voided

Sample Type

- A = Alpha spec
- F = fecal
- U = urine
- S = Start or stop date

(Note: For urinalysis, we always expect “A”, because all urinalyses were done this way since October 1989. The other entries are for database flexibility)

Reason – Reason for scheduling a sample:

- 1S = acute exposure
- 2S = follow-up sample to an acute exposure
- 3 = routine (chronic) exposure
- 2R = follow-up to routine (chronic) exposure
- 4 = background sample
- 5 = baseline sample
- 6 = random sample
- 8 = start date of exposure *
- 9 = stop date of exposure *
- 0 = acute exposure
- * (These should appear only in separate records introducing a series related to a chronic intake study).

Retention Class – Assigned retention class: ICRP-30 (1979) classes D, W, Y.

(Class Q indicates 90% Class “Super – W” (120 lung retention) and 10% Class Y, 8 µm AMAD.)

Volume (mL) or Weight (g) – sample volume (urine) or weight (fecal)

Total Uranium Result (dpm) - This is the normalized total uranium urine result in dpm/d

Type Assimilation

- H = inhalation
- I = ingestion
- J = injection

Incident Date – Date of documented radiological exposure incident, if any

Incident Time – Time of documented radiological exposure incident, if any

5A.1.3 *in vivo* Data

Dept – Department number of employee at the time of monitoring

Run Date – Date subject counted

Run No. – Laboratory identifier of counting session

Weight (lbs.) – Subject weight in pounds

Chest Thickness (in.) – Subject chest wall thickness in inches (May be in tenths of inches for some years)

Surface Contamination Code – A six-digit code indicating whether or not surface contamination was detected.

- If the first position is a '1', there was no surface contamination found, and the remaining 5 digits should be '00000'.
- If the first digit is a '2' or a '3', there was surface contamination found and the remaining 5 digits give the survey meter reading of the contamination.

Type Analysis – Code indicating the activity regions analyzed. There are two distinct sets of codes depending upon the date of the count.

From 10/62 – 1971, the codes are:

- 0 = Background run
- 1 = Uranium
- 2 = New hires/control counts
- 3 = Thorium
- 4 = Neptunium
- 5 = Miscellaneous
- 6 = Second thorium count

Since 1972, the codes are:

- 1 = ^{235}U
- 3 = Thorium
- 4 = ^{238}U and ^{235}U
- 6 = Thorium and ^{235}U (first thorium count)
- 7 = ^{235}U and Thorium (second thorium count)

Material Type – This code provides additional details for cases where Type-Analysis = 0, 1, 2, or 3

For Post-1972 counts where Type-Analysis = 0;

- 00 = Background run
- 01 = Sensitivity Check

For Type – Analysis = 1 (either pre-1972 or post –1972) or Type-Analysis = 2 (post 1972);

- 01 = Unknown
- 02 = Depleted uranium
- 03 = Depleted uranium
- 07 = Normal uranium
- 13 = Enriched uranium
- 19 = Enriched uranium

For Type –Analysis = 3 (either pre-1972 or post –1972)

- 00 = Thorium (first exam)
- 02 = Thorium (second or later exam)

U-235 (µg) – U-235 activity detected in chest cavity.

(If the count is invalid, an '&' appears as the first character.)

Thorium (mg) – Thorium-232 activity detected in chest cavity

U-238 (mg) – Th-234 activity detected in chest cavity

Tc-99 (μCi)- Tc-99 activity detected in chest cavity

Current Dept – Additional instance of employee department number used historically for reporting purposes.

5A.2 URANIUM SOLUBILITY IN THE LUNG

For a workplace as varied as Y-12, it is clear that no single solubility type or particle size distribution will apply to all workers. Furthermore, accurate assignment of the uranium lung clearance type to a given bioassay result was considered virtually impossible because of uncertainties regarding chemical form and limitations of the personnel-tracking system (Barber and Forest 1995, p. 669). Exposure to Type M material from 1948 to June 1998 appears to be the most likely absorption type. After June 1998, exposure to absorption Type S material is more likely. However, the absorption type may be based on the monitoring data and/or claimant favorable assumptions.

5A.3 IN VITRO DETECTION LIMITS

This section summarizes information developed in Section 5.2 in the main body of the Technical Basis Document. The tabulated values for urinalysis results represent laboratory detection limits, L_D , and do not include uncertainties introduced by sample collection or conversion from submitted volumes to daily void volumes. As noted in Section 5, detection limits for some historical techniques remain to be identified, and will be reported in subsequent revisions as available.

Table 5A-1. Uranium urinalysis detection limits.

Method	Years	Detection limit (mass)	Detection limit (activity)	Convention
Fluorometry	1950–9/1989	7 $\mu\text{g d}^{-1}$	11 dpm d^{-1}	
Gross alpha	1950–1964		47 dpm d^{-1}	L_D
Gross alpha	1965–9/1989		26 dpm d^{-1}	L_D
Alpha spectrometry	10/1989–present		0.15 dpm d^{-1}	L_D

Table 5A-2. Other *in vitro* detection limits.

Analyte	Method	Years	Detection limit (activity)
Tritium/HTO	Liquid scintillation	10/1988–present	2000 dpm d^{-1} *
Isotopic plutonium/M, S	Alpha spectrometry	10/1989–present	0.025 dpm/sample
²⁴¹ Am/M	Alpha spectrometry	10/1989–present	0.05 dpm/sample
²²⁸ Th/M, S	Alpha spectrometry	10/1989–present	0.15 dpm/sample
²³² Th/M, S	Alpha spectrometry	10/1989–present	0.07 dpm/sample
²³⁷ Np/M	Alpha spectrometry	10/1989–present	0.1 dpm/sample

*(estimate)

5A.4 *IN VIVO* DETECTION LIMITS

From the review of Y-12 documents and conversations with present and former Y-12 staff, the vast majority of site operations from 1943 to the present involved uranium in a variety of chemical forms and degree of enrichment. Thus, it is concluded that the primary internal radiation exposure to Y-12 workers was from uranium. However, the internal dosimetry program has included limited monitoring for Cs-137, Tc-99, thorium, plutonium, Ac-228, and tritium, among other radionuclides. There are difficulties in interpreting these measurement data that can not be resolved satisfactorily at this time. These issues should not have a large impact on the worker's internal dose because exposure to uranium is the source of the greatest part of the internal dose.

Although internal monitoring has existed from the earliest days, the data from these measurements are not available prior to 1950. Guidance for the period from 1948-1950 is provided but it is not feasible to provide any guidance for exposures prior to 1948. This will be provided in revisions to this TBD.

This section summarizes information developed in Section 5.3 in the main body of the Technical Basis Document. Detection limits for some historical techniques remain to be identified, and will be reported in subsequent revisions as available.

For dose reconstruction purposes and based on review of the Y-12 data, the U-235 and U-238 lung count detection thresholds are, respectively, assumed to be 130 µg and 13.5 mg through 1990. Beginning in 1991, the critical level or MDA is supplied with the individual results.

Table 5A-3. Reported *in vivo* detection limits for other radionuclides.

Radionuclide	Reported limit (nCi)	Reported limit (dpm)	Citation year
²³² Th/M, S	0.6	1300	1965
²³⁷ Np/M	2.7	5900	1961
²³⁷ Np/M— ²³³ Pa/M, S	0.255	560	1961
⁶⁰ Co/M, S	0.66	1500	1961
⁹⁵ Zr/M, S— ⁹⁵ Nb/M, S	1.0	2200	1961